



I HEREBY CERTIFY THAT THIS CORRESPONDENCE IS BEING DEPOSITED WITH THE UNITED STATES POSTAL SERVICE AS FIRST CLASS MAIL IN AN ENVELOPE ADDRESSED TO: COMMISSIONER FOR PATENTS, P.O. BOX 1450, ALEXANDRIA, VA 22313-1450, ON THE DATE INDICATED BELOW.

BY:

*Renee Conti*

Date:

*November 4, 2004*

**MAIL STOP AMENDMENT**

**IN THE UNITED STATES PATENT AND TRADEMARK OFFICE**

In Re Patent Application of:  
Atsushi Ueda et al.

Conf. No.: 7373

: Group Art Unit: 1745

Appln. No.: 10/058,707

: Examiner: Raymond Alejandro

Filing Date: January 28, 2002

: Attorney Docket No.: 10059-404US  
(P27007-01)

Title: Non-aqueous Electrolyte Secondary Battery

**SECOND DECLARATION OF ATSUSHI UEDA UNDER 37 C.F.R. § 1.132**

I, Atsushi Ueda, declare and state as follows:

1. I am a co-inventor of the invention described and claimed in the above-identified patent application. I am the same Atsushi Ueda who executed a first Declaration under 37 C.F.R. § 1.32, filed in this application on June 15, 2004.

2. I am familiar with the above-referenced application, and in particular with the Office Action dated July 7, 2004 (Paper No. 20040629). I am submitting the present Declaration to overcome the § 103(a) rejections of claims 1-16 by demonstrating the different effects observed when vinyl ethylene carbonate (VEC) is added to a cyclic carbonic acid ester having no carbon-carbon unsaturated bond (C) and to a solvent mixture containing a cyclic carboxylic acid ester (A) and a cyclic carbonic acid ester (C).

**The Examiner's Position and the Purpose of the Declaration**

3. The Examiner has taken the position that the closest prior art to the invention includes U.S. Patent Application Publication No. 2003/0118913 of Takami et al. ("Takami"),

U.S. Patent No. 6,315,918 of Mita et al. ("Mita"), EP 0 796 510 ("EP '510"), U.S. Patent Application Publication No. 2001/0018152 of Kida et al. ("Kida"), U.S. Patent No. 6,090,506 of Inoue et al. ("Inoue"), and U.S. Patent Application Publication No. 2002/0001756 of Hamamoto et al. ("Hamamoto").

4. In the Office Action dated July 7, 2004, the Examiner rejected claims 1-16 under 35 U.S.C. § 103(a) as being obvious over combinations of Takami and Mita in view of EP '510, Kida, Inoue and/or Hamamoto. The Examiner argued that all of the claimed elements are taught or suggested by the proposed combinations of cited references, including the inclusion of VEC to a solvent mixture containing a cyclic carboxylic acid ester (A) and a cyclic carbonic acid ester (C).

5. The purpose of this Declaration is to demonstrate that the addition of VEC to a solvent mixture containing components (A) and (C) results in a dramatic improvement in the initial charge/discharge efficiency of the resulting battery, an increase which is much greater than that observed by adding VEC to a solvent containing only component (C). Further, these data demonstrate the inferior initial charge/discharge efficiency exhibited by a battery in which the non-aqueous solvent contains a mixture of components (A) and (C) relative to a battery containing only component (C). These results would not be expected based on the prior art, and such a demonstration will thus overcome the rejections of claims 1- 16.

#### Background and Purpose of the Invention

6. The non-aqueous electrolyte secondary batteries according to the present invention were developed to overcome problems known in the art, such as deterioration of charge and discharge characteristics. The batteries according to the invention have excellent charge and discharge characteristics, particularly at low temperature, and show satisfactory charge and discharge characteristics, even after being exposed to high temperatures over a period of time.

7. According to the presently claimed invention of claims 1-15, non-aqueous electrolyte secondary batteries which exemplify these properties are realized by using a non-aqueous solvent containing: (A) a cyclic carboxylic acid ester; 0.5 to 20 volume % of (B) a cyclic carbonic acid ester having at least one carbon-carbon unsaturated bond and comprising vinyl ethylene carbonate (VEC); and (C) a cyclic carbonic acid ester having no carbon-carbon

unsaturated bond. Cyclic carboxylic acid esters (A) are prone to decomposition by hydrofluoric acid, possibly deteriorating the cycle characteristics of the battery. Additionally, the use of  $\text{LiBF}_4$  also deteriorates cycle characteristics of the battery because the degree of electrolytic dissociation of  $\text{LiBF}_4$  is small or a film formed on the surface of the negative electrode comprising graphite is weak. However, as shown in Tables 10 and 11 of the present application, the use of both salts in combination, particularly in the claimed ratio, improves cycle characteristics of the batteries and decreases the amount of gas generated, and also results in favorable capacity maintenance rates and cycle life. Further, the use of components (C) and VEC in combination with component (A) results in favorable battery characteristics which overcome these disadvantages.

8. In order to demonstrate the effect of including VEC in the non-aqueous solvent on the initial charge/discharge efficiencies, a series of batteries was prepared using different solvent mixtures containing components (C); (A) and (C); (C) and VEC; and (A) and (C) and VEC. The initial charge/discharge characteristics of these batteries were measured and compared.

#### Experimental Procedure

9. Batteries were produced using the procedure described in Example 1 of the above identified application. Non-aqueous electrolytes were prepared by dissolving  $\text{LiPF}_6$  in each of the following solvent mixtures at a  $\text{LiPF}_6$  concentration of 1.25 mol/L.

##### *Solvent Mixtures Used to Prepare Non-aqueous Electrolytes*

(EC= ethylene carbonate; PC = propylene carbonate; VEC = vinylethylene carbonate; GBL = gamma-butyrolactone)

- (a) EC and PC in a volume ratio of EC:PC of 80:20
- (b) EC, PC, and VEC in a volume ratio of EC:PC:VEC of 75:20:5
- (c) GBL, EC, and PC in a volume ratio of GBL:EC:PC of 50:40:10
- (d) GBL, EC, PC and VEC in a volume ratio of GBL:EC:PC:VEC of 50:35:10:5

10. As in Example 1, each of the four batteries was charged and discharged at a constant current of 300 mA at an ambient temperature of 20 C, with a final charging voltage of 4.1 V and a final discharge voltage of 3.0 V.

## Results and Discussion

11. The Table below shows the results obtained for the initial charge/discharge efficiency for each of the batteries, which was calculated as the percentage of the discharge capacity to the charge capacity at the final charging voltage and final discharging voltage.

12. As can be seen from comparing the data in the Table, battery (a), containing 80:20 EC:PC, had an initial charge/discharge efficiency of 80%, and battery (b), containing 75:20:5 EC:PC:VEC had an initial charge/discharge efficiency of 85%. The addition of VEC to a non-aqueous solvent containing only components (C) thus increased the initial charge/discharge efficiency by only 5%.

**Table**

<b>Battery</b>	<b>Composition of Non-Aqueous Solvent*</b>	<b>Initial Charge/Discharge Efficiency (%)</b>	<b>Effect</b>
(a)	EC 80: PC 20	80	
(b)	EC 75: PC 20: VEC 5	85	Addition of VEC to (a) + 5%
(c)	GBL 50: EC 40: PC 10	50	Addition of GBL to (a) -30%
(d)	GBL 50: EC 35: PC 10: VEC 5	83	Addition of VEC to (c) +33%

\* All percentages by volume

EC = ethylene carbonate; PC = propylene carbonate; GBL = gamma-butyrolactone; VEC = vinylethylene carbonate

13. However, battery (c), containing GBL, EC, and PC (50:40:10), exhibited an initial charge/discharge efficiency of 50%, whereas battery (d), prepared by adding VEC to the mixture of GBL, EC, and PC, exhibited a significantly increased initial charge/discharge efficiency of 83%. Accordingly, by adding VEC to the solvent mixture containing GBL, EC, and PC, the charge/discharge efficiency increased by 33%.

14. Further, by comparing batteries (a) and (c), it can be seen that addition of GBL (component (A)) to a non-aqueous solvent containing components (C) reduced the initial charge/discharge efficiency by a significant amount: 30%.

15. It can thus be concluded from the above Table that the addition of VEC to a solvent mixture containing a cyclic carboxylic acid ester ((A), here, GBL) and cyclic carbonic acid esters ((C), here, EC and PC) is much more effective at improving the initial charge/discharge efficiency than when VEC is added to a solvent containing only cyclic carbonic acid esters (C). Furthermore, the addition of VEC to a solvent containing components (A) and

(C) overcomes the reduced initial charge/discharge efficiency which is caused by inclusion of component (A).

Conclusion

16. This Declaration demonstrates that by preparing non-aqueous electrolyte secondary batteries according to the invention, which contain VEC in combination with a cyclic carboxylic acid ester (A) and a cyclic carbonic acid ester (C) having no carbon-carbon unsaturated bond, favorable and unexpected results are obtained, particularly with regard to initial charge/discharge efficiency. None of the prior art of record specifically teaches the use of VEC in combination with component (A) and (C) in the non-aqueous solvent, nor makes obvious the results which have been observed by the present invention.

I declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further, that those statements were made with the knowledge that willful false statements the like so made are punishable by fine or imprisonment, or both, under Section 1003 of Title 18 of the United States Code; and that such willful statements may jeopardize the validity of the application or any patent issuing thereon.

Dated: October, 26, 2004

Atsushi Ueda

Atsushi Ueda